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THE NATURE OF VARIOUS ASPECTS OF LUMINESCENCE OF
PHOSPHORS WITH DEEP LOCALIZATION LEVELS

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26 May 1947

[All figures are appended.]

The characteristics of the flash, phosphorescence, and luminescence illumination on excitation of $\text{CaS}:\text{SrS}-\text{Ca}$ and La -phosphors. The elementary processes of which these forms of illumination consist are explained, and the relationship of their intensities in various phases of luminescence is established.

(Presented at the luminescence colloquy at the Physical Institute, Academy of Sciences USSR, 19 February 1947.)

Introduction

1. Purpose of Research

A new type of luminescent compound, which can accumulate and preserve a large quantity of excitation energy for a considerable period, was developed in 1941-1945 in the Physical Institute, Academy of Sciences USSR. These compounds release the energy only upon exposure to infrared rays or visible light. The new compounds are alkaline-earth phosphors (CaS - SrS and SrS), activated by a combination of two or more rare earths (Ca , Sm and Eu , Eu).

The optical characteristics of the phosphors: the spectral composition of the radiation, the position of the absorption spectra and the luminescence output of fully excited phosphors, in relation to the radiations emitted, were investigated in this research. The role of the component parts of the phosphors, the basic material and the two activators, in the appearance

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of the flash was also explained. It appeared that the absorption spectra and the flashes were independent of the basic material, but were determined chiefly by Ce and Eu, while Sm is the activator of the flash and increases its intensity many times. At the same time a similar project was conducted in the US, about which information is now being published in US journals (2-6). The results of US authors basically coincide with the results obtained at the Physical Institute, Academy of Sciences USSR, and pertain to the same questions--the establishment of the role of the individual components of the compounds and explaining the conditions of the existence of an intense flash.

To understand the flash processes it is necessary, however, to make a detailed study of all the types of luminescence which occur in phosphors of a given type. The present work aims: (a) to explain the relationships between luminescence from the infrared rays, the phosphorescence occurring after removal of thermal excitation and luminescence at the moment of excitation and (b) to ascertain the nature of these processes. The conclusions reached on the nature of the various processes, to a certain extent, can be qualitatively applied to other phosphors with deep localization levels. On the other hand, the numerical values given below for the individual quantities undoubtedly depend considerably on the composition of the phosphor and the method of its preparation. Nevertheless, the correct relationships between the basic materials determining the course of luminescence in actual phosphors are of fundamental theoretical importance, since even an approximate indication of these relationships makes it possible to judge the validity of some theoretical presentations and the fallibility of others.

2. Investigation of Compounds

The object of investigation was a layer of $\text{CaS}:\text{SrS}-\text{Ce}$, Sm, La-phosphor of thickness ~ 0.07 mm. The layer was excited throughout its thickness and readily permitted the passage of infrared radiations. During radiation, it had approximately the same brightness from both sides. The luminescent light was green, which corresponds to Ce radiation. The phosphorescent light was a yellow-green, slightly different from the light of the flash. The presence of La increased the flash somewhat, but had no noticeable effect on the other characteristics of the phosphors.

The scheme of the arrangement is given in Figure 1. The phosphor Ph was enclosed between two quartz plates on stand E. L_1 is a mercury lamp FPK-2 used for excitation; F_1 is Shott's UG 11 light filter which passes lines 366, 334, 312, and 303 m μ . The distance from lamp to phosphor was ~ 30 cm. The exciting ultraviolet stream incident on the phosphor was equal to $3 \cdot 10^{-3}$ W/sq cm. This excitation is considered normal. It was weakened up to 200 times by screen N_1 ; the use of the lens L_1 increased it 15-25 times. Thus the total possible variation in excitation was as much as 5,000 times the lowest excitation value in different tests. The length of excitation time varied from 0.04 seconds to 30 minutes, i.e., 45,000 times.

For experiments on the study of the action of individual lines of the mercury spectrum, the supplementary lamp L_2 was used. The radiation of the excited phosphor was produced by incandescent lamp L_2 , 300 W, covered with a black Shott filter, UG 6, which allowed the passage of all infrared rays beginning with 850 m μ . The minimum distance of the phosphor from the lamp was 28 cm. The energy irradiation density of the phosphor surface was $\sim 10^{-2}$ W/sq cm, which later was taken as a unit of radiation. Lamp L_2 moved along scale S; in addition, its illuminating beam was varied by screen N_2 . The brightness of the phosphor illumination was measured by a visual photometer. The brightness of the source for comparison was varied by the optical wedge D. The uniformity of all lamps was controlled by measuring devices. The temperature at the test was 24°C. (The mechanism was prepared by Z. A. Trapeznikov.)

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3. Method of Measuring

A complete group of measurements consisted of investigating the increase of phosphor luminescence at excitation, the decrease in phosphorescence after removal of excitation, and the brightness of luminescence for various phases of radiation by infrared rays. Integration under the areas of the intensity curves for all the indicated luminescences give the so-called "light sum" [area under curve] of these processes.

During radiation, lamp L₂ was first placed on the end of scale S, and the brightness of the initial phosphor flash was measured. Then the lamp was quickly raised (region a, Figure 2) until a position was reached at which phosphorescence registered was maximum brightness on the photometer. At succeeding approach of lamp L₂, this brightness of illumination was maintained until further approach of the lamp and the phosphorus was impossible. Further, the decay of the phosphor luminescence was measured at a constant distance of lamp L₂ from phosphor Ph.

A. Basic Characteristics of Luminescence During Excitation

1. Flash of CaS·SrS-Ce, Sm, La-Phosphor

Three processes of luminescence and the nature of the radiation of CaS·SrS-Ce, Sm, La-phosphor. An examination of Figure 2 establishes the relationship between the three processes of phosphor luminescence: luminescence due to excitation (fluorescence), phosphorescence, and flash. Since the increase of the brightness of luminescence during excitation by a light of normal intensity occurs very quickly, a very weak activating light was used to make this transition apparent. The length of excitation time (30 minutes) was such that the "light sum" of the flash had time to reach a value close to the maximum ($S=0.96 S_{\text{max}}$).

It follows from Figure 2 that the luminescence during excitation does not immediately attain its levelling-off value, but shows a certain amount of inertia. The drop in luminescence after removal of exciting radiation occurs almost immediately. For an initial qualitative orientation in the energy relationships of these separate processes, we notice that the "light sum" of luminescence during excitation for the first 160 seconds, in which there occurred a noticeable increase of luminescence, was approximately 11,500 units, while during this time there was accumulated 2,600 units of the flash "light sum" and only 50 units of the phosphorescent "light sum." Thus, the average excitation energy in the indicated time interval was distributed between the enumerated processes in the ratios 100:22.6:0.43.

In order to explain the characteristics of the examined phosphors, which are sensitive to infrared and visible rays and have the property of conserving for a long time the light sum of excitation, a hypothesis was put forward by the author in 1942 (7) concerning the existence in this type of phosphor of two systems of localization levels, relatively shallow and deep. To an electron, which has fallen into the conductivity level, there is presented the possibility of immediately recombining, joining an ionized center descending to one of the inner levels. From here, with further thermal agitation, the electron can again be raised to the conductivity level and upon its recombination give phosphorescent luminescence, or it can finally descend into a deep localization level. From here the electron can be freed only by the action of infrared rays or by strongly heating the phosphor. This hypothesis, founded on general considerations and comparisons of the characteristics of phosphors having flash properties with the properties of the ordinary self-radiating phosphors, will be submitted to direct experimental examination in the present paper. The data given below substantiate its validity.

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Of the three processes: luminescence during excitation, phosphorescence, and flash, only phosphorescence is a simple process. As will be shown below, at the flash there occurs a partial transfer of electrons from the deep level to the phosphorescent levels, and luminescence of secondary phosphorescence is added as a consequence of the flash. During excitation, luminescence is even more complex. This luminescence is basically brought about by the recombination of electrons from the conductivity level without prior localizations, or after localizations on shallow level in which the electron can remain for only a small part of a second. On this recombining luminescence, phosphorescence is superimposed. In addition to this, in theory there can exist and be added to the first two, (a) flash luminescence due to the excitation rays themselves and (b) monomolecular luminescence due directly to excited ions (8). It is shown below that the first two processes undoubtedly exist and make up the basic part of luminescence during excitation. The last two in the described phosphors under our experimental conditions did not play a significant role.

2. Inertia of Luminescence During Excitation

The question of lag, or inertia, in the formation and disappearance of luminescence, observed during excitation of phosphor, always has a great significance in decisions as to its nature. From the viewpoint of the concepts examined above, it would be expected that in the first moments of excitation of electrons, which had fallen in the conductivity level, they should in theory be directed to the local levels of the flash and phosphorescence, which are completely empty at that moment. Only an insignificant proportion will recombine with ions of the first electrons to fall into the conductivity level, since, during the first moments of excitation, the number of ions is very small.

Depending on the increase in the number of ions determined by the number of localized electrons, and the decrease of the number of unfilled local levels, the number of recombinations will also increase and the number of localizations will decrease. The increase of luminescence at activation must have a certain inertia. An instantaneous rise of luminescence would indicate either the presence of a considerable amount of the usual fluorescence, i.e., direct monomolecular excitation and radiation of ions of the activator, or extremely little likelihood of localization. As a consequence, recombination, even with a very small number of ions, would occur faster than the fall of the electrons on many free levels of capture.

At greater intensities of the exciting light, the filling of the flash and phosphorescence levels and the formation of a large number of ionized centers naturally occurs at a rapid rate and the luminescence reaches maximum brightness very quickly. Because of this, to find lag or inertia in the increase of luminescence, small intensities of excitation were used: $I_E = 0.05$ and $I_F = 0.01$ of the normal. In Figure 3 the growth of luminescence is depicted at these excitations. The inertia of the rise is clearly distinguished. Even after 5 minutes of radiation, the brightness of luminescence has not yet reached its limiting value. The first readings give values of brightness which corresponds only to ~30 percent of the final values of brightness. It follows that the small-inertia processes (fluorescence) can not be significant. The rise of luminescence is basically represented by a scheme corresponding to a picture of gradual filling of the localization levels.

In accordance with the same concepts, the drop of luminescence after removal of excitation should occur very quickly since the outer electrons, at the moment of removing excitation, should recombine or be localized very quickly. Experiments substantiate this expectation. In Figure 2, it is

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clear that, at the moment of stopping excitation, an intermittent decrease occurs in the brightness of luminescence. Here very weak excitation was used at which the brightness of luminescence during excitation was only 12 times greater than the brightness of phosphorescence at the time of the first reading. During excitation of normal intensity, the luminescence drop, at the moment of stopping the excitation, occurs still more abruptly.

It is possible to suppose that all the luminescence during excitation is phosphorescence, the drop of which, in the first moment just after stopping excitation, occurs so fast that it cannot be investigated without special phosphorescopic devices. This opinion is wrong. It can easily be shown that if by phosphorescence is meant a process caused by electrons being freed by light from sufficiently deep localization levels, then the luminescence at the moment of excitation will be connected with it only to a certain extent (usually small). Actually, as the brightness of the exciting light is increased, the brightness of luminescence during excitation also grows. It does not often display a tendency towards saturation, and in many phosphors can in practice be made as large as desired. The phosphorescence, on the other hand, in a series of cases, reaches saturation relatively easily and does not change, even with further increase of intensity of excitation. Thus, the phosphor luminescence at excitation is not exhausted by phosphorescence alone.

3. Basic Characteristics of the Flash

a. Proportionality of Brightness of Flash and Intensity of Radiation.

During study of the flash, first of all it was shown that the brightness of the flash, other things being equal, is proportional to the amount of radiated light falling on the phosphor. The intensity of the flash was 50 times the incident light. The average error of individual measurements reached 6 percent. Experimental data is given in Figure 4.

This result was used later in the investigation of the "light sums" of the flash.

b. Flash Inertia

In the case of a rather great intensity of radiation, the drop in brightness of the flash occurs so fast that the impression produced is that of a practically instantaneous flash, with a very steep drop in brightness which commences at the very beginning of luminescence. To eliminate the abrupt drop in luminescence of the flash, it was studied under very weak radiation. In Figure 5, the initial phases are depicted of radiation of various excited phosphors at 1/300 of the value of normal radiation. In both cases we see that, in spite of the small intensity of infrared light used, the very first readings give almost the maximum value of the flash. Thus, the test substantiates the fact that basically, for visual observations, the flash is without lag.

This was to be expected if we consider that the flash appears as the result of the direct recombination of electrons, raised by infrared rays from deep localization levels into the conductivity level. There are, however, weak maxima on both curves. They are formed comparatively slowly. Thus, on the upper curve, the increase in brightness of the flash continued for 2.5 minutes. These maxima can be explained as a second, very inert, supplementary process, namely, secondary phosphorescence which arises as the result of secondary localization on phosphorescence levels of part of the electrons which had been raised by infrared rays from deep levels into the conductivity level. (For further details, see below.)

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4. Equivalence of Time and Excitation Intensity for Filling Flash Levels

Since, in the test arrangements, the escape of the electrons from the deep flash levels without the aid of infrared rays was excluded, it was to be expected that the filling of the flash levels is determined by the product $I_B t$ (the intensity of excitation I_B by the time t of excitation) and not by each of these quantities separately.

The test completely substantiated this opinion. There was obtained, for example:

Table 1

I_B	t	$I_B t$	S_{BC}	I_B	t	$I_B t$	S_{BC}
1.0	1.0	1.0	4,900	1.00	0.5	0.5	4,320
0.2	5.0	1.0	4,925	0.01	50.0	0.5	4,450

This rule was later given even wider verification. This, however, cannot be extended to very small intensities since it will then be so large that it becomes impossible to disregard the natural (temperature) decrease of the "light sums" of the flash.

5. Laws of Increase of "Light Sums." Laws of Decay of Flash and Phosphorescence

A similar examination of these processes constitutes a special report. Here we will limit ourselves to the remark that their course can be satisfactorily described by Becquerel's formula: $I = (a + bt)^{-\alpha}$, or even the simplified formula: $I = At^{-\alpha}$. Our new tests fully substantiated this rule both in regard to the flash and to the phosphorescence. The growth of the latter in the described phosphors must occur somewhat differently from the usual phosphor manner since here there are two different systems of local levels. The formula $I = At^{-\alpha}$ was verified, nevertheless. For α in the majority of cases, a value somewhat less than unity is obtained.

B. Phosphorescence Levels and Flash Levels

1. Distinction Between Phosphorescence and Flash Levels

The simplest presentation of the nature of the flash is that it arises from the speeding up of the luminescence of a phosphor, the brightness of whose natural luminescence has fallen below the limit of sensitivity of the eye. From this viewpoint there is only one system of electronic levels, common to phosphorescence and to flash. The invalidity of this representation is evident from the relation of the "light sums" of both processes (see Figure 2). The "residual" process--the flash--surpasses many times the "basic" process--phosphorescence. The very large value of the "light sums" of the flash persists even after sweeping the excited phosphor for several days, although the residual phosphorescence will decrease sharply after a prolonged period. A no less convincing objection against the indicated simplified treatment of the flash is that many phosphors, having a bright and slowly decaying phosphorescence, do not have the characteristics of the flash or thermoluminescence. These considerations compelled the author, in 1942, to adopt the above indicated concept of the existence of two localization levels in the phosphor in question, - shallow phosphorescence levels and deep flash levels (7). In the present paragraph, a description of tests was attempted to establish the difference between the characteristics of the flash and phosphorescence levels which come chiefly from the difference of their depths. The question of the number of these and other levels and of the relative probability of their being filled is examined below.

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2. Function of Intensity

The ratio between flash and phosphorescence "light sums" is a function of the intensity of the excitation light. The suggested great difference in the depths of the flash and phosphorescence levels should be reflected in the thermal stability of the electrons localizing on them. On the flash levels, the electrons remain for a very long time; on the phosphorescence level, on the other hand, they break out very quickly due to thermal agitation. Therefore, during prolonged action of exciting rays on the phosphorescence levels, a dynamic equilibrium is established between the electrons which are coming and going. For a given intensity of excitation, the ultimate value of the "light sum" of phosphorescence S_{ϕ} is established after a period not exceeding the period of full natural decay of phosphorescence (in our case ~ 5 minutes). Its size is decreased with the decrease in excitation intensity.

On the other hand, the electrons which are localized on the flash levels do not break out into the conductivity level. The "light sum" of the flash $S_{\phi c}$ continues to grow even after the establishment of the equilibrium on the phosphorescence levels. Because of this, during a lengthy excitation, the ratio of the flash "light sum" to the phosphorescence "light sum" must increase as the excitation intensity is decreased. The experiment carried out verified this proposition. Excitation lasted for 10 minutes during which time the full values of the "light sums" of phosphorescence were established for the given excitation intensity. The excitation intensity I_B had a two hundredfold variation from the normal to 0.05 times the normal. The ratio of the "light sum" $S_{\phi c}$ (flash) and S_{ϕ} (phosphorescence) were changed from 22.5 to 146. Figure 5 is a graph of this ratio as a function of $\log I_B$.

It must be noticed that the flash "light sums," corresponding to an excitation of 10 minutes, were not the maximum, and that at an excitation of 0.005 of the normal excitation intensity, the flash "light sum" had time to reach only 33 percent of its maximum value.

Table 2 gives ratios of the "light sums" of phosphorescence S_{ϕ} to the flash "light sum": $S_{\phi c}$ (maximum) and $S_{\phi c}$ (after 10 minutes of excitation) at various intensities of the exciting light I_B .

Table 2

The Ratios $S_{\phi} / S_{\phi c}$ and $S_{\phi} / S_{\phi c \text{ MAX}}$ as a Function of I_B

I_B	$\frac{S_{\phi}}{S_{\phi c}}$	1	0.2	0.05	0.01	0.005
$S_{\phi} / S_{\phi c}$		0.020	0.022	0.017	0.011	0.007
$S_{\phi} / S_{\phi c \text{ MAX}}$	0.040	0.029	0.021	0.011	0.005	0.002

3. Dissimilar Effects of Infrared Rays on Electrons in Levels of Flash and Phosphorescence

The difference between phosphorescence and flash levels is also manifested by the different effects of infrared rays on the "light sums" of the two processes.

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The experiment was then repeated three times with simultaneous action of excitation and infrared rays of various intensities: 0.2, 0.5 and 1.0 times the normal. The "light sums" of phosphorescence S_{ϕ} and of the flash $S_{\phi\phi}$ decreased in different amounts. The phosphorescence proved less sensitive to infrared rays than the flash. Figure 7 gives the corresponding relationships. The experiment further showed that increasing excitation intensity decreases the effect of radiation of the infrared rays of the previous intensity, since the infrared rays of this intensity do not have time, even to former extent, to radiate to localization levels, which are rapidly being filled.

4. Secondary Phosphorescence During Flash

A very graphic demonstration of the separate existence of phosphorescence and flash levels is secondary phosphorescence which arises as the result of the action of infrared rays on which phosphor had already radiated phosphorescence. The decayed phosphorescence, after a short action by infrared rays, is renewed and decays again, and on repeated exposure to infrared rays appears again. The process can be repeated dozens of times. The explanation is that the electrons of the flash levels, falling into the conductivity level under the action of infrared rays, do not all recombine, but a certain number undergo repeated localization, partly on free-flash levels, partly on levels of phosphorescence. The latter also give secondary phosphorescence. The kinetics of the process is examined in detail by the author in a separate paper. Only a qualitative description of this phenomenon is presented here, such as the existence of localization levels and the rise of phosphorescence.

5. Radiation and Freezing of Phosphorescence

The simplest method of determining approximately the depths of the localization levels consists in determining the temperature at which the radiation of the corresponding level takes place. To find the depth of the levels of phosphorescence, it is necessary to excite the phosphor at the freezing temperature of phosphorescence and then gradually heat it to determine the radiation of the localization levels. The experiments carried out showed that, in phosphor excited at -183°C , a significant sharply decaying phosphorescence differs considerably in color from the usual decaying phosphorescence. Investigation of the variation of the quantity $\Delta S_{\phi} / \Delta t^{\circ}$ where S_{ϕ} is the light sum accumulated by the phosphor and t° is temperature, discloses the existence of special levels of phosphorescence which radiate in the approximate temperature region -100 to -30°C .

Similar experiments on thermal radiation of phosphors excited at low temperatures have recently been carried out by Z. L. Morgenshtern, whose results agree with ours. (15)

6. Relation of Number of Phosphorescence and Flash Levels and Probabilities of Their Being Filled

To solve the problem of the relative probability of the transition of electrons from the conductivity level to the phosphorescence and flash levels, the ratio of the "light sums" of both processes was studied for increasing exposures. At the same time the exciting intensity was taken at a maximum and the length of the exposure was made so small that the radiation of the phosphorescence in the time of excitation could be disregarded. Numerical data is assembled in Table 3.

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Table 3

Comparison of Rate of Filling of Flash and Phosphorescence Levels During Intensive Excitation

Length of Activation in Sec	0.01	0.04	0.10	0.20	0.50	1.00	3.00	300
Flash Sum S_{FC}	162	750	1980	2840	4320	4980	6200	6580
Phosphorescence Sum S_{P}		40	95	136	202	300	410	343
Ratio $S_{\text{FC}}/S_{\text{P}}$		19	20.8	20.9	21.4	16.6	15.1	19.2

The average ratio $S_{\text{FC}} : S_{\text{P}} \approx 19.4$. As can be seen from the table, the ratio of the phosphorescence and flash "light sums" in filling their levels remains unchanged within the limits of error of measurement. The constancy of this ratio was substantiated by a second method, Figure 8. The first points on Figure 8 correspond to small exposure periods of 0.04; 0.2 and 1.0 second. The last point in Figure 8 and the last row in Table 3 correspond to excitation lasting 300 seconds. Obtaining the previous value of the ratio of phosphorescence and flash even for these prolonged exposures showed that the excitation light used was so intense that it filled the phosphorescence levels much faster than their temperature radiation.

It followed, from the graphs showing the variation of flash and phosphorescence "light sums" with exposure periods used in the previous cases, that the "light sums" of both processes could be considered as maximums, i.e., for the given condition of the exciting light, the flash as well as the phosphorescence levels were filled. Because of this, the ratio of their "light sums" immediately gives the relative number of filled levels. Since the ratio of the flash and phosphorescence "light sums" remain unchanged in all ranges of the filling of these levels, it must be recognized that their filling occurs uniformly, i.e., the probability of electron localization on both systems of levels is alike.

We must also notice that the data in the table characterizes a full store of "light sums" of the deep levels, since the infrared rays radiate the flash to the end. The radiation of phosphorescence occurs without our thermal participation and continues under various filling conditions of the deep levels. Owing to the constancy of the ratio of the "light sums" of both processes, it happens that the differences in the degree of filling the deep levels do not indicate the radiation of phosphorescence. The successive electron transitions occurring in this process in the deep flash levels, or the percentage of these transitions at all stages of filling the deep levels, evidently can be disregarded.

The ratio $S_{\text{P}} : S_{\text{FC}} \approx 0.05$; i.e., the number of phosphorescence levels is only 5 percent of the number of flash levels, if we assume that the freeing of the electrons from the shallow levels of phosphorescence is not accompanied by losses occurring at thermal radiation of flash "light sums" (9). If these losses also occur at phosphorescence, then the number of phosphorescence levels should be 25-30 percent of the number of the flash levels.

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3. The Nature of Luminescence During Excitation

1. Two Representations of Luminescence During Excitation

In section "A" above, the basic characteristics of luminescence which are found in excitation were described: the lag or inertia of formation, the very great value of the "light sums" radiated at these processes, and at visual observation the very abrupt and practically instantaneous drop of luminescence after removal of excitation, which becomes normally decaying phosphorescence only to a small extent. At the basic examination of luminescence characteristics during excitation, it was assumed that the basic part of it brings about the immediate recombination of the electrons from the conduction level which do not have a previous localization on planes of a considerable size.

However, there exists another point of view for this process. Lenard (10) found that in certain cases the exciting rays can produce radiation. In 1938 Gernsey and Mott (11) ascribed the radiation of phosphorescence to this agency. This viewpoint was supported by Ritt (12). In 1943 this representation of radiation of the action of the exciting rays was used by V.V. Antonov-Romanovsky (13) to explain the rapid drop of luminescence of silicate phosphor and later of a group of phosphor examined by us (14).

According to this view, the intensity of luminescence, at the moment of excitation, is determined by the number of electrons raised by the exciting rays themselves from the localization levels. The maximum luminescence during excitation is established when equilibrium is attained between the freeing and localization of the electrons. In the development of this hypothesis in detail, it can be assumed either that the radiation occurs here, as for infrared rays, through $\beta\alpha$ or that the exciting rays are not directly incident on the localized electrons.

The existence of radiation action of the exciting rays in certain cases was proved. However, as will be clear from the experiments described below, in the phosphors investigated by us, it cannot make up a significant part of the luminescence during excitation.

2. Nature of Variation

The variation in the brightness of luminescence at the moment of excitation and the variation in the brightness of flash in relation to the number of localized electrons, are of a completely different nature.

From the view of the hypothesis of the radiation action of exciting rays, luminescence during excitation is the same as flash, merely coming as a result of the exciting light itself. Thus the brightness of luminescence during excitation must increase with increase in the number of electrons localized on the flash levels, according to the rule explaining the increase in brightness of the flash. Both curves were measured independently, i.e., the increase of the brightness of luminescence at the moment of excitation was studied for successive increases in flash "light sums," and the brightness of the flashes were measured for various values of the "light sums."

The tests were conducted with the intensity of the exciting light equal to 0.05 times the normal. To determine the dependence of the flash brightness on the size of the "light sums," the excitation was stopped in a determined moment. The initial flash brightness and the flash "light sum" were determined after the phosphorescence radiation. Figure 9 shows the curves for the increase of the "light sums" of excitation (curve I_M) and the increase of flashes (curve $I_{\phi\phi}$) versus the magnitude of the flash "light sums." These curves are distinguished from one another not only quantitatively but also qualitatively, since they have different curvatures.

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In estimating what part of the luminescence during excitation can come from the radiation of the exciting rays on the flash levels, it must be kept in mind that the curve of this luminescence has a form represented on Figure 9 for I_{pg} . On the other hand, the curve for the direct recombination of the electrons must rise continuously. Therefore, corresponding to the process of direct recombination, the difference of curve I_M and the curve of radiation by exciting rays, cannot have a maximum value. It follows from this that the luminescence brought about by the radiation action of exciting rays will be less than 0.1 of I_M in all cases.

3. Intense Luminescence

Intense luminescence during excitation in certain cases can arise at very small flash "light sums." In this connection the action of the mercury line 435 m μ is interesting. Table 4 includes the values of the relative brightness of luminescence I_M caused by the action of lines 435 and 366 m μ and the flash "light sums" S_{pg} which were obtained by the action of each line separately and together.

Table 4

λ (m μ)	435	366	435 + 366
I_M	312	168	—
S_{pg}	2	223	196

We see that, in the experimental arrangements, when the line 435 m μ gave off a greater brightness of I_M than the line 366 m μ , this luminescence occurred at very small flash "light sums." It could have been assumed that the greater brightness of I_M at the presence of only a small "light sum" S_{pg} arises due to the unusually strong radiation action of line 435 m μ . However, the direct determination of the radiation action of line 435 m μ by comparing the "light sums" obtained at the action of one line 366 m μ and the two lines 366 + 435 m μ together, showed that the radiation action of line 435 m μ was small.

The nature of luminescence during excitation for the line 435 m μ is not yet completely established. It is possible that, apart from recombinations from the conductivity levels, there exists a direct excitation of cerium ions.

The considerations mentioned make us ascribe the chief part of the luminescence during excitation to the recombination of electrons straight from the conductivity levels or of electrons undergoing localization in very shallow levels.

4. Distribution of Electrons From Conductivity Level and Absolute Number of Transition Electrons

After examining the general picture of illumination of SrS-Ce, Ba, La-phosphors, the question naturally arises as to the distribution of electrons of the conductivity level between the localization level and ionized centers. The general amount of energy transformed by a phosphor in a unit of time, if diverted from extinguishing processes, can be determined by the strength of luminescence during excitation after this luminescence, as a result of filling localization levels, reaches its full development. This energy, calculated per unit of excitation energy, was determined for various energies of excitation from many tests. The obtained results fluctuated within the following limits:

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I_B	15	1	0.2	0.05	0.01	0.005	Average
$6 \cdot 10^{-3} I_{M00}$	9.45	9.34	8.37	7.79	6.42	8.52	8.38

knowing the rate of growth of the "light sum" S_{PC} of flash and the "light sum" S_{ϕ} . Further, as a function of exposure $\theta = I_B t$, it is easy to compute $\Delta S_{PC} / \Delta t$ and $\Delta S_{\phi} / \Delta t$, increments of energy needed in various phases of exposure to fill both systems of local levels. The subtraction of both values $\Delta S_{PC} / \Delta t$ and $\Delta S_{\phi} / \Delta t$ from the full energy of luminescence during excitation obviously gives the curve of the growth of this luminescence according to the exposure increase. The rate of all curves is represented on Figure 10: Σ is the level of full luminescence at the moment of excitation; $\Delta S_{PC} / \Delta t$ is curve of the filling of the flash levels; $\Delta S_{\phi} / \Delta t$ is curve of the filling of the phosphorescence levels; and I_B is curve of the growth of luminescence during excitation according to the exposure increase.

This curve is obtained by subtracting curves $\Delta S_{PC} / \Delta t$ and $\Delta S_{\phi} / \Delta t$ from Σ . Its growth on transformation of coordinate θ to t is very close to the experimentally obtained growth of the luminescence at activation shown in Figure 3. It follows from Figure 10 that at the beginning of exposure, and also in later stages of flash radiation, there are very considerable quotas of electrons from the conductivity level which are directed to localization levels.

We must notice that the raising of electrons by infrared rays from the localization levels to the conductivity level is not accompanied by strong extinguishing, since during a steady cycle, the brightness of luminescence at the moment of excitation was similar to that when the exciting light alone was acting, as well as when it acted in conjunction with infrared rays. In the first case, the electrons recombine immediately from the conductivity level. In the second case, part of the electrons had previously passed through the localization levels; the flash "light sums," as was shown above at the simultaneous action of infrared rays, are smaller.

Knowing the absolute value of the flash "light sums," in units of apo-stilb/minute, (Stilb is USSR unit of brightness) of our phosphor the number of localized electrons could be computed approximately during full excitation of the phosphor. It was of a smaller order than the number of Sm atoms.

Conclusions

The fundamental results of the present research are:

1. Establishing the separate existence in CaS:SmS-Je , Sm, La-phosphors of deep flash levels and shallower phosphorescence levels.
2. Finding the uniform filling of both systems of levels, i.e., the equal probability of electron localization from the conductivity level on the levels of both system.
3. Establishing the nature of the basic part of luminescence during excitation as luminescence of the recombination of electrons which either have not undergone previous localization or have been localized on very shallow levels.
4. Establishing the rate-of-change probability of the recombination and localization processes with increase of exposure and filling of the localization levels.

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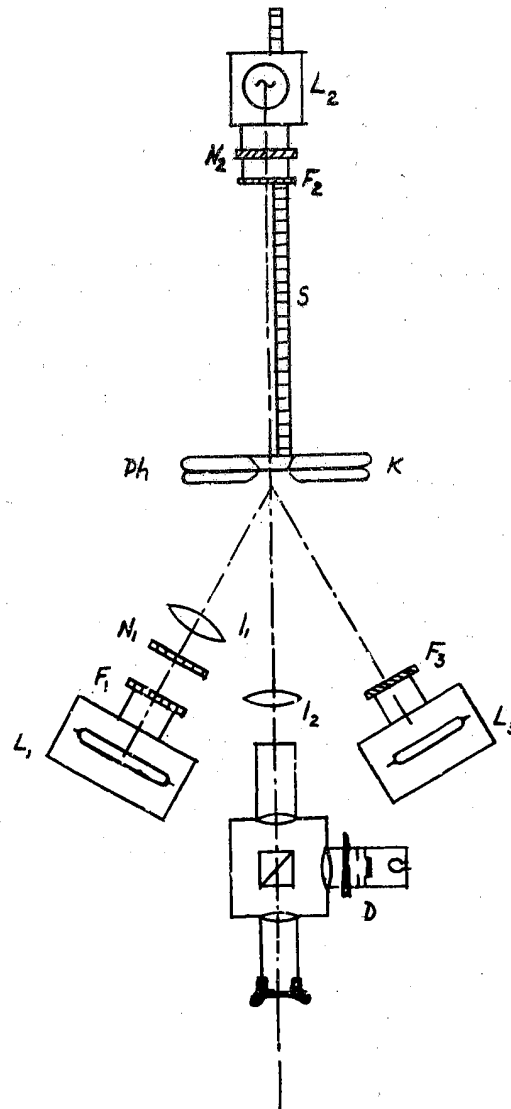


Figure 1

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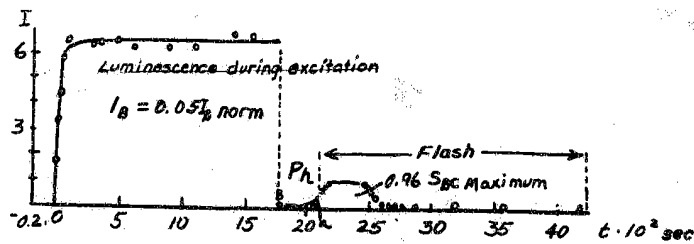


Figure 2

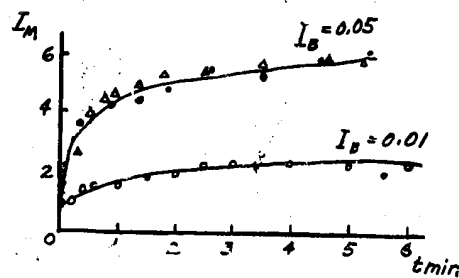
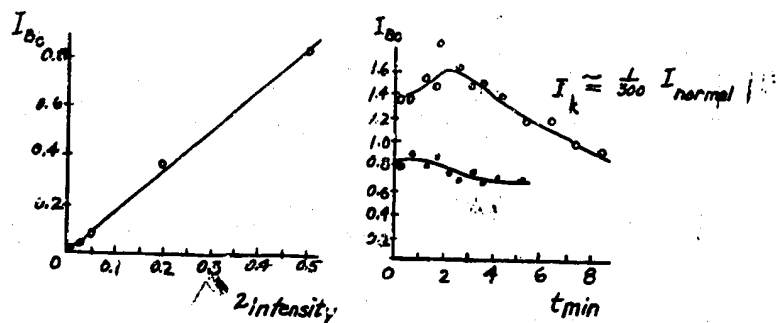


Figure 3



Note: BC means flash

Figure 4

Figure 5

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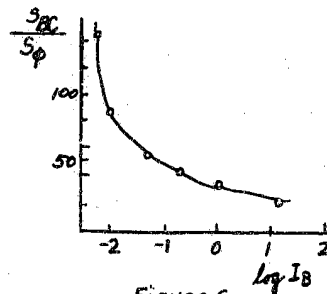


Figure 6

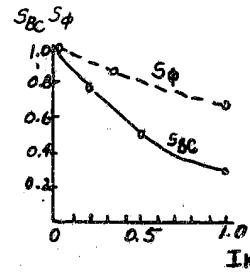


Figure 7

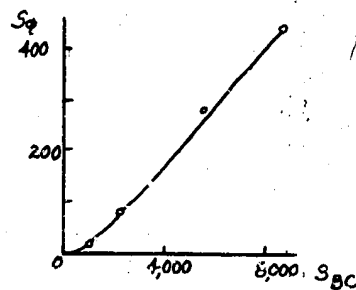


Figure 8

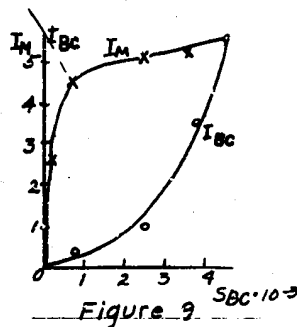


Figure 9

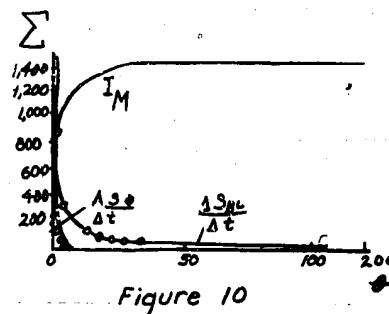


Figure 10

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